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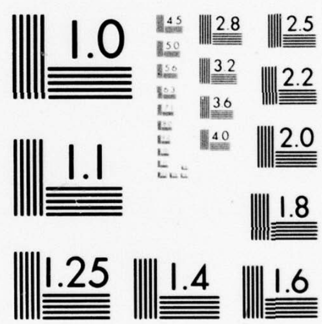
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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

The generation of short far infrared pulses by using a mode-locked CO_2 pump laser is explored experimentally and theoretically. It is demonstrated that pulses of <1 nsec are obtainable from CH_3F at 496 μm and D_2O at 385 μm. Theoretical estimates indicate the possibility of pulses in the 300-600 psec range with a conversion efficiency comparable to the Manley-Rowe limit.

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I. Introduction

This final report summarizes research performed under contract DAAG29-77-C-0025 during the period 1 August 1977 to January 20, 1979. The topics involved in this contract were techniques for generating temporally short far-infrared (FIR) or near millimeter wave pulses and equipment purchases to upgrade our experimental studies involving FIR lasers, materials and components. The remainder of this report is devoted to the details of these topics.

II. Equipment Purchases

In the development of relatively primitive spectral regions such as the FIR, one frequently encounters questions dealing with the linear properties of materials, components, etc. along with the usual problems associated with transient detection characteristic of the pulsed operational mode of optically pumped or electrically pumped FIR sources. Problem areas such as these are not indigenous to the FIR, but have plagued researchers in the area for some time, including ourselves.

Two recent acquisitions will help alleviate these problems but also create a new one. These are the acquisition of a Fourier transform spectrometer (Beckman RIIC IR-720M) and a transient digitizer (Biomation #6500). These units will considerable expand our capability in material and component

studies and in our ability to capture, digitize and store transient signals.

To further enhance the capability of these units, it was decided to use the equipment allocation of this grant to construct a small computer, the equivalent of a PDP-11. The capabilities of this system will allow us to do spectral scans on the IR-720M over a wavelength range and resolution which exceed the onboard computer memory of that unit and to control, acquire, average and data massage digitized pulses from the 6500 unit. In addition, future acquisition plans call for a higher speed digitizer (Tektronix R7912AD) which can also be controlled by the PDP-11 based system. Finally, the system will allow 'in-house' computation with line printer and graphics capability.

The computer system, constructed on campus, is LSI-11 based and is comprised of the hardware items listed in Table I. These items permit handling of up to 32 kwords (16 bit) of fast memory and 512 kwords of slow memory, vector graphics with 750×1024 point resolution, digital graphing and line printing. The onboard software includes the RT-11 V3B operating system, FORTRAN V3, MACRO, the equivalent of CALCOMP graphics routines and other assorted utility programs. An interface/averager board has been constructed to allow data acquisition and control of the #6500 system and is in the process of being debugged. Output from the IR-720M is in digital

Table I. Computer System Breakdown.

SYSTEM	ITEM	VENDOR	MODEL	SPECIAL FEATURES
I/O	Graphics Terminal	Tektronix	4006	750×1024 pt. resolution
	Line Printer	Digital Equipment	LA36	300 baud
	Digital Plotter	Houston	HiPlot	.005" resolution on 8½×11"
Main Frame	CPU	Digital Equipment	KD-11-HA	LSI-11
	Back Plane	MDB	BPA-11	9 quad slots
	Extended Arith.	Digital Equipment	KEV-11	
	Serial I/O	Digital Equipment	DLV-11-J	4 ports
	Ram	Monolithic	MSC4604	32k×18 bit
Mass Memory	Floppy Disc	Advanced Electronic Design	6200F-A	quad disc drive, dual density, 256 kwords/disc

form requiring only a FFT program for handling. This powerful system will be a welcome addition to our experimental efforts, considerably enhancing our existing instruments as well as contributing its own special features.

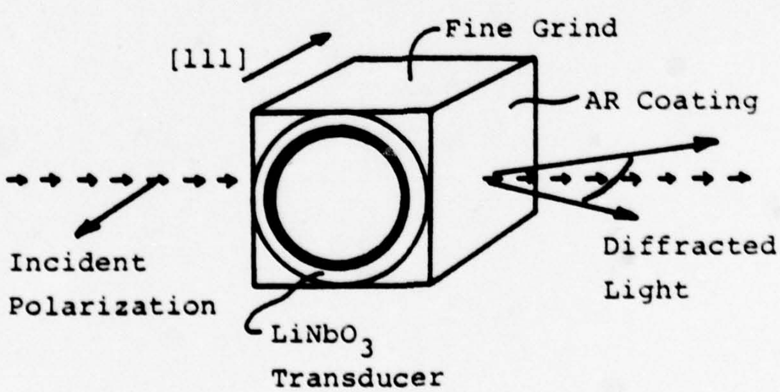
III. Short Pulse Generation

A major portion of the contract effort was devoted towards generating and characterizing short FIR pulses from optically pumped systems. The motivation of this work was to seek ways to produce FIR pulse powers which exceed those obtained by conventional techniques of optical pumping, with the added constraint of not increasing the size of the pump. Since the Manley-Rowe limit governs the energy conversion of the system, then to generate pulses which would seemingly violate this limit when applied to power would require some effective pulse compression schemes. Of the possible interactions which could lead to pulse compression (stimulated backward wave Raman emission, superradiance and synchronous-mode locked pumping), synchronous mode-locked pumping was chosen because of the flexibility and control afforded by the approach and because of the demonstrated spectacular success in visible pumped dye laser systems.[1,2] Our initial observations and findings have been reported and are reproduced in Appendix I and II.[3,4] In what follows, further experimental and theoretical details will be outlined.

a. Experiment

The experiment was comprised of a mode-locked TEA CO_2 laser and either a single-pass or cavity FIR cell. The CO_2 laser was of conventional geometry and is further described in Appendix II. Only supplemental information will be presented here.

The mode locker was a 20 mm cube Ge acousto-optic modulator, custom made for the experiment by Laser Optics and shown below. The sound entrance and exit faces [111] were optically polished flat and parallel to standard CO_2 etalon specifications and the CO_2 exit and entrance faces had an additional AR coating set for 9.6 μm. The optical polarization was in the [111] direction, the direction of the sound wave propagation.



Mode-Locker Geometry

The sound transducer was custom fabricated by Valpey-Fischer with the following identifications:

> $LiNbO_3$ Transducer: 36° rotated, y-cut, 21 MHz, fundamental, 20 mm dia., polished, c/gold coax, w/5/8" spot.

The transducer is mounted on the Ge cube with a thin layer of Nonaq stopcock grease (Fisher Scientific) which is water soluble. The grease thickness determines the acoustic match and hence the transducer impedance. The transducer is driven by $\sim$ 5W RF at 21 MHz (half the cavity frequency) from a pulsed (msec) RF source. No special handling or operational problems were noted except the ease with which the AR coatings damaged.

The reaminder of the experimental system is documented in Appendix II.

The findings of the experiment were very encouraging in that equivalent mode-locked FIR pulse trains were generated with individual pulse widths of $<$ 1 nsec. Similar results have been reported recently on an equivalent system and set limits on the pulse widths from CH_3F at 496 μm, $\lesssim$ 355 psec and from D_2O at 385 μm, $\lesssim$ 710 psec.[5] In addition, FIR powers in the range of 10-100 kW were detected which are greater than or equal to the Manley-Rowe limited conversion of the average, i.e., unlocked and single mode, pump power.[3,5] The significance of these results are twofold: first, since the FIR pulse is approximately a factor-of-ten shorter than the inverse line-

width, the effective gain-bandwidth needed to account for the pulse width must be associated with near-resonant stimulated Raman emission off each pump frequency component and second, the effective compression associated with mode-locking the pump is producing a compressed output with powers equal to or exceeding the Manley-Rowe limited average power, thus demonstrating the utility of the approach. Further experiments are in order involving larger cells and involving linear or non-linear correlation techniques for better short pulse estimates.

b. Theory

Paralleling the experimental investigation was a theoretical investigation needed to provide insight into the conversion dynamics. This was necessary because of the transient or sub-T_2 interaction time scale, placing the experimental conditions outside the range of previous short pulse situations.[6,7,8,9,10]

The theoretical approach was tackled from four standpoints: 1) weak pump, single frequency FIR probe, CW pump train, 2) weak pump with a single pulse, space-time growth, 3) strong pump, spatial steady state, and 4) strong pump, space-time growth. In the first approach, we assume a weak CW pump and a tunable single-frequency FIR probe wave. The normalized gain is then solved for and should illustrate the

Raman contributions. This is indeed the case as can be seen in Fig. 3 of Appendix II. Here are shown two gain profiles corresponding to two different detuning conditions for the pump. Clearly seen in this figure are the individual Raman contributions due to each frequency component of the pump, illustrating the main contention of the experiment. The resulting solution is also listed in Appendix II, Eq. (5) along with some variable definitions used in Fig. 3.

In approach 2), we switch from a CW train to a single pump pulse mainly to deduce the spatial rate of growth of the FIR and some tentative time behavior. The details are also listed in Appendix II with the main result being that the FIR evolves as $\exp[\frac{2}{3}(\alpha x)^{\frac{1}{2}}]$ where x is a normalized space variable and $\alpha = LG\tau^3$ with GL some constants and τ is retarded time measured from the start of the assumed square pump pulse. This unusual spatial growth is reminiscent of a 'lethargic' gain amplifier[11] and growth in the normal (not resonantly enhanced) Raman regime on a transient basis.[9,10] In addition, because of the exponential time dependence of $\tau^{3/2}$, one might expect Stokes pulses of shorter duration than the pump pulse.

The next approach, 3), was more realistic in the sense that it closely parallels the experiment differing only in the steady state assumption, and some minor conditions. Assuming that the pump and Stokes center frequencies are on line center, the normalized equations of motion for the

off-diagonal and diagonal density matrix elements and the Stokes field growth are:

Off-Diagonal:

$$\dot{R}_{13}(M) = -\mathcal{E}_p f(M)[N_1(M)-N_3(M)]-\mathcal{E}_s f(M)R_{12}(M)-R_{13}(M)/T_2$$

$$\dot{R}_{12}(M) = \mathcal{E}_s f(M)R_{13}(M)-\mathcal{E}_p f(M)R_{12}(M)-R_{12}(M)/T_2$$

$$\dot{R}_{32}(M) = -\mathcal{E}_s f(M)[N_3(M)-N_2(M)]+\mathcal{E}_p f(M)R_{12}(M)-R_{32}(M)/T_2$$

Diagonal or Population:

$$\dot{N}_1(M) = 2\mathcal{E}_p f(M)R_{13}(M)-[N_1(M)-N_1^e(M)]/T_1$$

$$\dot{N}_2(M) = -2\mathcal{E}_s f(M)R_{32}(M)-N_2(M)/T_1$$

$$\dot{N}_3(M) = 2[\mathcal{E}_s f(M)R_{32}(M)-\mathcal{E}_p f(M)R_{13}(M)]-N_3(M)/T_1$$

Field Growth:

$$\partial\mathcal{E}_s/\partial z = -G\sum_M f(M)R_{32}(M) - \kappa\mathcal{E}_s$$

where $\mathcal{E}_p = \mu_{IR}E_p/2\hbar$, $\mathcal{E}_s = \mu_{PER}E_s/2\hbar$ with μ_{IR} equal to the band moment ($\sim$ .1D) and μ_{PER} equal to the permanent dipole moment ($\sim$ 1D) and N_1^e is the equilibrium population in the ground state. (The energy level configuration is in Appendix II, Theory Section). Also assumed are simple R-branch transitions for the pump and Stokes fields with both fields orientated along the z-quantization axis for which the M dependent matrix element is:

$$f(M) = \left\{ \frac{(J+1)^2 - M^2}{(2J+1)(2J+3)} \right\}^{\frac{1}{2}}$$

with J the lower level rotational quantum number.[11] The parameter G is equal to $k_s \mu_{PER}{}^2 / 4\hbar\varepsilon_o$ and κ is some assumed field loss term, i.e., diffraction or absorption with a value 10^{-3}-$10^{-2}cm^{-1}$.

These equations were solved in the so-called 'steady-state' regime, obtained by setting $\partial E_s/\partial z = 0$ which would occur in a very long interaction cell with a space invariant pump.[12] The pump was chosen to have a parabolic power spectra (see Appendix II, Fig. 3a) in a CW train with pulse period 25 nsec. The amplitude was abitrary and J was set equal to 5 to approximately reflect the D_2O situation. The equations were solved using a simple one-step Rung-Kutta method with a field initial condition of $\mathcal{E}_s = 1$ or $I_s \sim 10^{-15}$ W/cm^2 corresponding to 300°K blackbody at $\sim$ 500 μm in a $\sim$ 100 MHz bandwidth.

The results of the calculation yielded a periodic FIR output as expected except that the output did not stabilize until the second - fourth pump pulse, see Fig. 1. This implies that the residual effects of the previous pulse (FIR plus pump) are affecting the buildup of the next pulse and it is not until a few cycles have transpired before the initializing conditions stabilize. Similar features have been found recently for self-induced transparency by CW trains.[13] This also implies that short FIR pulse generation by a single pump pulse

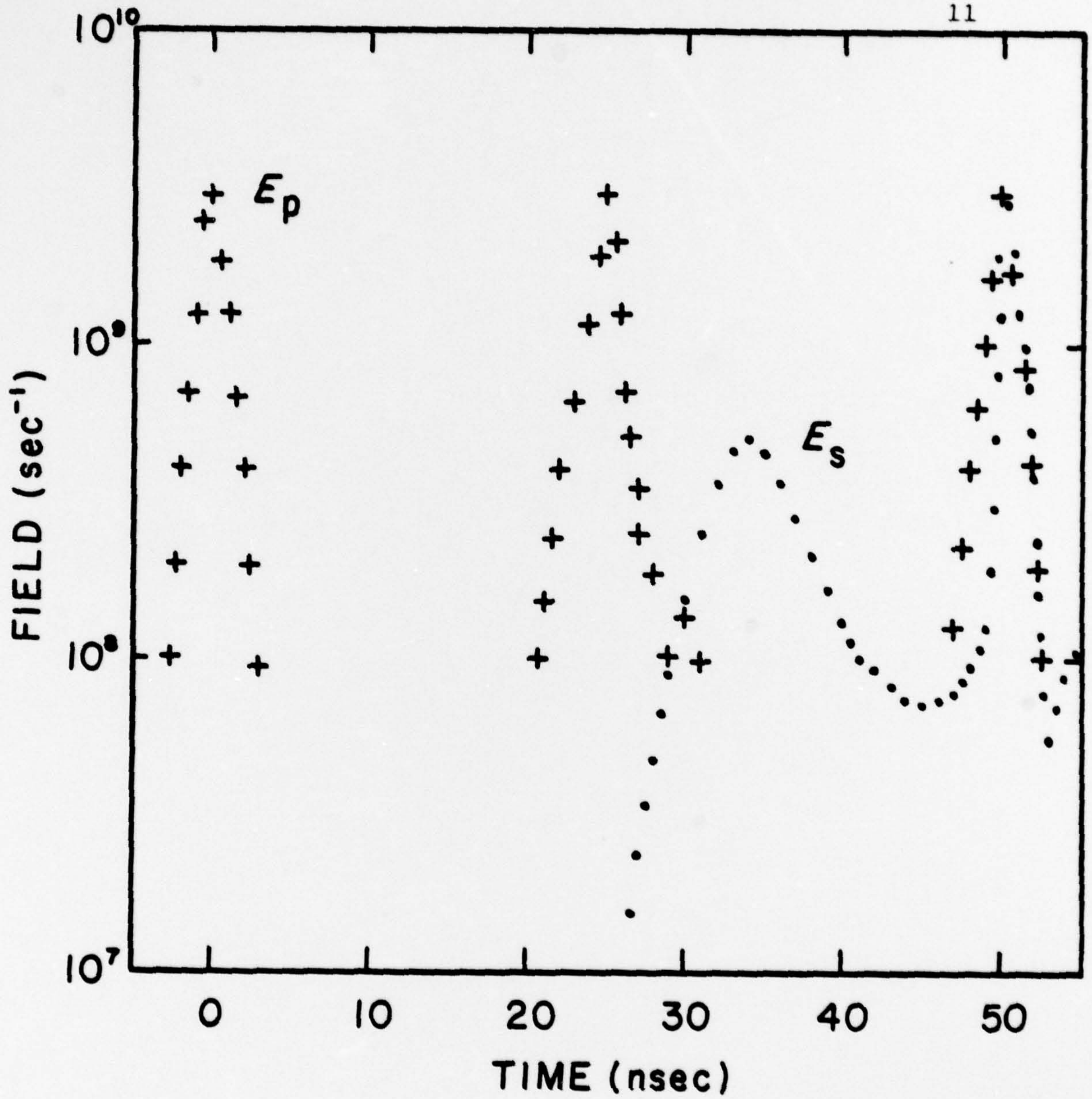


Figure 1. Normalized fields ($\mathcal{E} = \mu E/2\hbar$) versus time for $GN_1/\kappa = 2\times10^{10}$. The calculation was started at $t = -3$ nsec with an initial condition of $\mathcal{E}_s = 1$. For clarity only $\mathcal{E}_p > 10^8$ are shown. For $t > 50$ nsec, the system has stabilized. The pump field is approximately 1 MW/cm^2, 1.2 nsec duration.

may not be particularly rewarding.

The calculations also show that the FIR pulse is similar in shape to the pump; that the FIR pulse may, depending on conditions such as G/κ, occur during or after the pump pulse; that the duration of the FIR pulse may be less than ½ that of the pump and that FIR fields (E_s) about equal to pumping fields (E_p) are generated. The latter implies FIR power densities of $>$ 10 kW/cm^2 with durations in the range $\sim$ 300-600 psec for mode locked pump pulses of $\sim$ 600-1200 psec.

While these calculations are far from exhaustive in the range of parameters and conditions studied, the results of the calculation are in reasonable accord with experiments even though the latter are probably not in the steady state. The calculation also oversimplifies the CH_3F situation which is comprised of a K-multiplet of which components K = 1-6 are thought excitable by the pump.[14,15]

The fourth calculational approach entails the full space-time solution of the previous equations. This approach has not been undertaken yet primarily because of the expense of running propagation codes of this type. However from our previous studies on superradiant systems, we anticipate that the steady state may be well approximated somewhere in the range $1 < \kappa z \lesssim 10$ or $z \sim 1$ - 10m; interaction lengths which are within experimental range.[16]

c. Summary

In concluding, it appears that with the use of a mode-locked pump, FIR pulses of duration $<T_2/10$ can be generated. The results of a theoretical investigation show that the exact pulse duration is controlled by the pumping pulse duration (which itself can be controlled with the use of cavity filters) with probably pulse durations $\sim$ 300-600 psec. Anticipated FIR power densities of $\sim$ 1% of the pumping density are indicated in the steady state which is close to the Manley-Rowe limit. The spatial conversion distance has not been found but is expected to be in the range 1-10m. The next logical step would be a further compression to stack the generated FIR pulse perhaps gaining another factor-of-ten in peak intensity as well as obtaining a single pulse.

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IV. Scientific Personnel Associated with Grant

1.	Paul D. Coleman	Principle Investigator
2.	Thomas A. DeTemple	Principle Investigator
3.	L. T. Specht	Research Assistant
4.	S. H. Lee	Research Assistant

V. Thesis and Publications Associated with Grant

1. S. H. Lee, "Short FIR Pulse Generation", M.S. Thesis, Electrical Engineering, 1979.
2. S. H. Lee, "Synchronous Mode Locked Pumping of Low Pressure Gases", Optics Letters, 4, 6-8 (1979).
3. T. A. DeTemple, "Picosecond Far Infrared Pulses: Some Approaches", in Proceedings of the International Conference on Lasers '78, (V. Corcoran, editor), SPS Press, McLean, Virginia, 1979, pp. 104-110.

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Synchronous, mode-locked pumping of gas lasers

Appendix I.

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The generation of mode-locked far-infrared pulses by synchronous, mode-locked optical pumping by CO_2 lasers is demonstrated for the first time. The gain bandwidth implied by the observed short pulses (<1 nsec) exceeds the homogeneous linewidth of 40 MHz (T_2 = 8 nsec) under the experimental conditions and is attributed to an effective inhomogeneous broadening in molecules with a K-multiplet structure such as $^{12}CH_3F$ and also to the presence of near-resonant stimulated Raman emission from each frequency component of the pump. This implies that the duration of pulses generated in low-pressure gases by this technique is determined by the greater of the pumping-pulse bandwidth or the linewidth of the lasing transition.

Laser optical pumping is a powerful technique used for the generation of new laser frequencies from gases, liquids, and solids with demonstrated success spanning the range from the ultraviolet to the millimeter. In addition to the generation of new frequencies, the approach also offers flexibility in the time domain with the use of a Q-switched or mode-locked pump. For example, mode-locked argon lasers have been used to pump dye lasers synchronously, resulting in mode-locked pulses from the latter with durations considerably shorter than from the former.[1] However, the linewidths available in condensed media are generally much larger than in the gas phase, suggesting that mode-locked pumping may not result in particularly short pulses from low-pressure samples. It is the purpose of this Letter to demonstrate mode-locked synchronous pumping of the low-pressure gases and to demonstrate that, as a consequence of near-resonant stimulated Raman emission, the pulse width from such systems is related more to the bandwidth of the pump than to the linewidth of the transition.

A convenient system for investigation is the CO_2-pumped far-infrared (FIR) laser, primarily because of the large gain available on these pure rotational transitions.[2] A mode-locked CO_2 TEA laser and a FIR waveguide laser were used in the experiment. The CO_2-laser cavity was 3.6 m long, grating tuned with an intracavity Ge acousto-optic mode locker of conventional geometry, and produced ~50-mJ, 5-MW peak power.[3] The FIR cavity consisted of an internal Au-coated flat mirror and an external, translatable Si etalon separated from the mirror by 3.6 m. The input end of the 40-mm i.d. Pyrex FIR waveguide was terminated with a Si Brewster window whose interior surface was coated with a multilayer dielectric ir mirror, transparent in the FIR.[4] The pump was reflected off this mirror and down the waveguide axis toward the internal mirror, making one round trip before being lost. A tungsten-wire-on-nickel MOM diode in conjunction with a 350-MHz (Tektronix 485) oscilloscope was used to detect the FIR; a photon drag detector was used for ir detection.[5]

With approximately 5-W pulsed rf drive to the modulator, reliable mode locking was obtained for the pump, an example of which is shown in Fig. 1(a) for the $P_9(20)$ line. In Fig. 1(b) are shown several trains observed from $^{12}CH_3F$ at 496 μm. (Because of photographic writing-rate limitations, the photographs were back illuminated and retouched with dots for reproduction purposes.) Similar trains were also observed from D_2O at 385 μm pumped by the $R_9(22)$ line, an example of which is shown in Fig. 1(c). The FIR power was estimated to be between 1 and 10 kW for both lines.

On an expanded time scale, individual pulses can be resolved as shown in Fig. 2. The impulse response of the oscilloscope, Fig. 2(a), was 800 psec (FWHM), while the shortest pulses from $^{12}CH_3F$ at 496 μm, Fig. 2(b), and D_2O at 385 μm, Fig. 2(c), ranged from 1.25 to 1.5 nsec, which was the approximate range of the shortest pump pulses shown in Fig. 2(d). Incomplete mode locking resulted in broadened pulses, examples of which are also shown. True pulse widths between 500 and 1000 psec were inferred for the shortest observed pulses by using a simple convolution of the oscilloscope im-

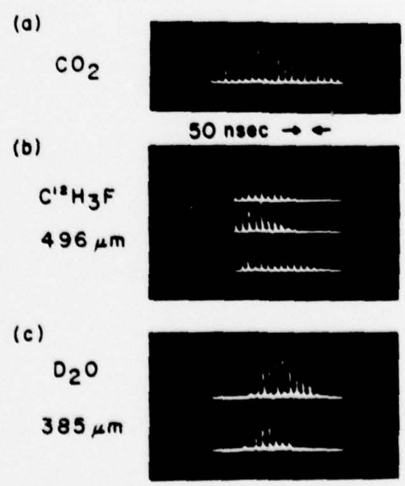

Fig. 1. Mode-locked pulse trains from (b) and (c) a synchronously pumped FIR waveguide laser and (a) from the pump. The pressure in the waveguide laser was 1 Torr for (b) and (c).

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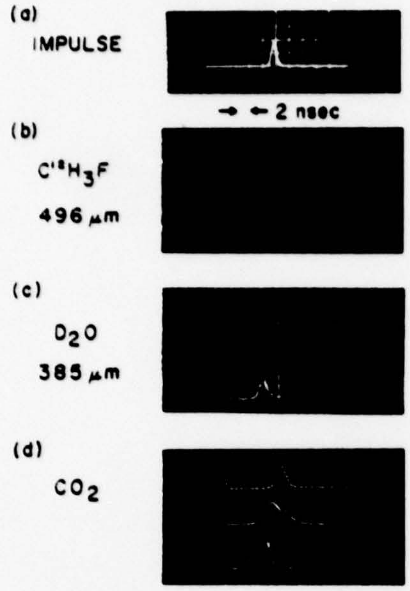

Fig. 2. Single pulses from the trains in Fig. 1 showing irregularities due to incomplete mode locking and normal locking. The impulse for (a) was a 400-psec triangular pulse.

Thus, since the actual pulse shapes and detector speed are unknown, we can only claim that the true pulse widths are <1 nsec.

It is of interest to note that, for the pressure conditions of Fig. 2, ~1 Torr, the pressure-broadened linewidth for both FIR transitions is 40 MHz, which, applying conventional wisdom, should yield a mode-locked pulse of 25-nsec duration, clearly much larger than what is observed.[6] For the case of $^{12}CH_3F$, the ir transition, ν_3 $^QQ(12,K)$, consists of a K multiplet of which components $K = 1$–6 are thought to be excitable by the ~1-GHz bandwidth pump.[7] The subsequent FIR laser emission from these multiplets would have a spectral spread of ~500 MHz, which could result in a mode-locked pulse of ~2 nsec, close to what is observed in Fig. 2.[8] For D_2O, the absorption and emission is an isolated transition which, if the emitted FIR were solely due to stimulated emission, should result in much longer mode-locked pulses than are observed.[9] However, near-resonant stimulated Raman emission can in effect considerably increase the gain bandwidth, resulting in much shorter pulses.

There is a growing body of evidence that stimulated Raman emission is present and important in many cw and pulsed systems, being solely responsible for the generation of many FIR lines.[10–13] Conceptually, it is also possible to have a stimulated Stokes wave corresponding to each frequency component of a multimode pump, implying that the emitted linewidth should be related to the pump linewidth. To illustrate this, a perturbation solution of the two-wave three-level system has been developed, assuming for simplicity a cw mode-locked pump-pulse train and a tunable single-frequency FIR probe wave. The resulting Stokes gain, G_s, is expressed as

$$G_s(y,\tau) \sim -\mathrm{Im}\sum_m\sum_n \times \frac{P_m P_n \exp[i(x_m - x_n)]\tau}{L(x_n - D)L(x_n - D - y)L^*(y + x_m - x_n)},$$

where x_n, y, and D are the detunings for the nth pump mode from pump-line center, the probe FIR from FIR laser-line center, and the pump-line center from ir absorption-line center, respectively, all in units of homogeneous halfwidths, and P_n and τ are the normalized pump-mode amplitudes and time (τ [illegible] T is the pulse period). The complex lineshape is defined as $L(z) = z + i$. In Fig. 3(a) is shown a synthetic parabolic power spectrum roughly approximating our pump, which would result in a mode-locked pulse of 1.2-nsec duration. Also shown on the same scale is a Lorentzian, which would be the characteristic lineshape of a FIR transition. In Figs. 3(b) and 3(c) are shown the normalized Raman gain profiles at the temporal peak of the pump for the power spectrum of Fig. 3(a), clearly showing the individual gain contributions due to each mode of the pump. The large contribution near $y = 0$ is due to a fully resonant Raman effect associated with the pump modes nearest ir line center. The two cases correspond to the $^QQ(12,2)$ transition in $^{12}CH_3F$ ($D = 2$) and to the 5_0–4_0 transition in D_2O ($D = 16$) responsible for the 385-μm line. In both cases, the gain

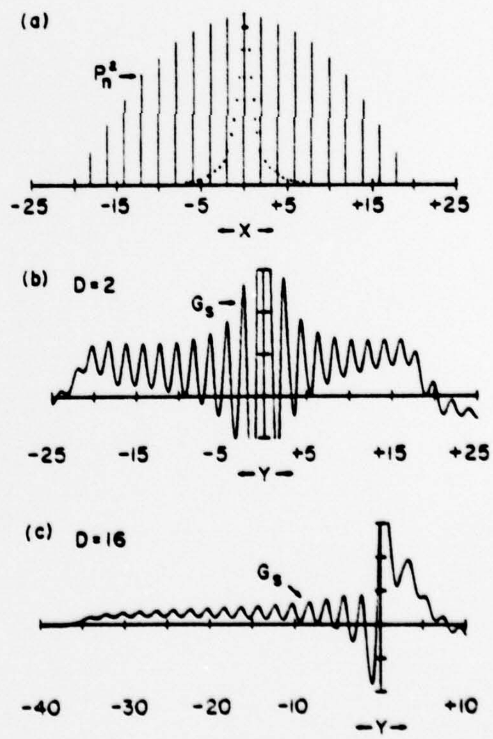

Fig. 3. (a) Synthetic power spectrum approximating the pump. (b) and (c) are small-signal Stokes gain profiles for the pump in (a); x, y, and D are normalized detuning parameters, which, for the conditions of Fig. 1, have a value of 1 unit = 20 MHz. The origin $y = 0$ is centered over the pump mode at $x = D$.

correspond to the ${}^{Q}Q(12,2)$ transition in $^{12}CH_3F$ (D = 2) and to the 5_0–4_0 transition in D_2O (D = 16) responsible for the 385-μm line. In both cases, the gain bandwidth, which persists only for the duration of the pump, is considerably larger than the bandwidth of the Lorentzian in Fig. 3(a), characteristic of a laser transition, which is the main feature to be illustrated by this simple treatment.

In conclusion, we have demonstrated that mode-locked synchronous pumping of low-pressure gases can result in a short-duration mode-locked output. The duration of the pulses is significantly shorter than the inverse bandwidth of the laser transition, a fact caused by an equivalent inhomogeneous broadening from systems with a K-multiplet structure or by stimulated Raman emission from each frequency component of the pump; a combination of both may also be present.[13] This implies that the pulse duration is determined by the greater of either the pulse bandwidth or the laser linewidth, the latter being the limit of, for example, dye lasers and the former being one possible limit for low-pressure gases.[1] The theoretical limit of the technique is not known, requiring a more-detailed analysis including saturation, ac Stark shifts, transient nutation effects, and dispersion in the FIR.[14] However, we remark that tunable stimulated Raman emission from pure rotational transitions in HCl has been observed over a 2-cm^{-1} tuning range, which could result in ~20-psec pulses with the use of a mode-locked pump.[15] With the existence of these short FIR pulses, new correlation techniques will have to be developed to permit more accurate estimates of the pulse duration. Finally, the approach may also be applied to other optically pumped gas lasers, such as the metal and halide vapor molecules in the visible, or to the CO_2-pumped midinfrared systems.

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Appendix II.

PICOSECOND FAR INFRARED PULSES: SOME APPROACHES*

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Abstract

Some possible approaches towards the generation of short far infrared pulses are discussed. In particular, synchronous, mode-locked pumping of far infrared lasers by CO_2 lasers is demonstrated to produce far infrared pulses with a time duration which is a factor-of-20 shorter than the inverse linewidth. This observation is attributed to either an equivalent inhomogeneous broadening in systems with a K multiplet structure or to stimulated Raman emission from each frequency component of the pump: a combination of both may exist. These imply that the ultimate pulse duration is determined by the greater of the pumping pulse bandwidth or the effective linewidth.

Introduction

The techniques which may result in the generation of short optical pulses may be grouped into two catagories: internal (to the source) and external generation. The latter is characterized, for example, by some shuttering mechanism; the AC Kerr or Duguay shutter being one used in the visible[1] and optically induced free carrier absorption/reflection being another used in the infrared.[2,3] The former is exemplified by various mode locking techniques, forced mode locking and synchronous mode locked pumping are two common approches, and by interactions which result in a self-compression such as superradiance[4] and backward stimulated Raman emission.[5]

Regarding the far infrared (FIR) spectral region which is loosely bounded between 50 μm and 1 mm, the approaches towards generating short FIR pulses are less well defined because of known laser and material limitations. For example, conventional wisdom argues that the pulse duration from a mode locked laser is proportional to the inverse linewidth[6] which, for typical operating pressures of around 1 torr, is approximately 40 MHz yielding pulses some tens of nanoseconds long in the FIR. Similarly, only free carrier absorption in semiconductors emerges as a viable external approach for shuttering because of the presence of background absorption in most candidate nonlinear materials.

It is the purpose of this paper to discuss short FIR pulse generation by one approach, synchronous mode locked pumping, which has been recently demonstrated in the FIR,[7] and to speculate on short pulse generation by optically induced free carrier absorption and by superradiance/backward stimulated Raman emission. In Section II, the experiment and results are presented while Section III continues a brief outline of some theoretical features of the approach. Section IV speculates on the other approaches while the results are summarized in Section V.

Experiment

Synchronous mode locked pumping, having been spectacularly demonstrated in dye laser systems,[8,9] is a natural approach to investigate since many of the present FIR sources are excited by CO_2 lasers. In addition, stimulated Raman emission has been identified as being the mechanism responsible for the generation of many FIR lines by optical pumping[10,11] suggesting that the effective bandwidth in these systems may be dominated by the pump bandwidth, rather than the transition linewidth, due to a stimulated Raman contribution from each frequency component of the pump. As a test of this possibility, an experiment utilizing a mode locked pump was initiated.

A diagram of the experiment is shown in Figure 1. The pump was a CO_2 TEA laser, grating tuned with an intracavity Ge acousto-optic mode-locker of conventional geometry.[12] In operation, the mode-locker was gated on a few msec before the TEA laser discharge in order to establish a good acoustic standing wave in the crystal. Since the mode-locker appears as a loss in the cavity, the resulting output energy is reduced somewhat from the unlocked case. With this system, we generated mode locked pules of ~ 50 mJ, composed of a train of ~ 1 nsec pulses with ~ 5 MW peak power.

The FIR cavity was comprised of an internal Au coated flat mirror and an external, translatable Si etalon. The overall cavity length was set equal to the TEA laser cavity

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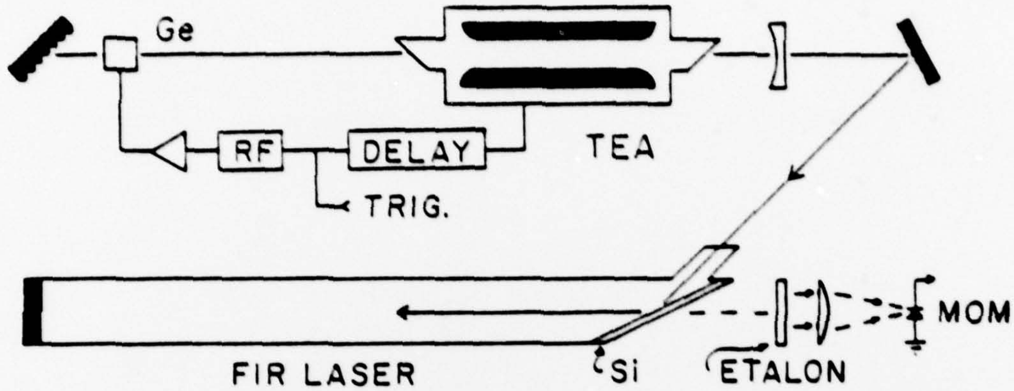


Fig. 1. Synchronously pumped FIR laser experiment.

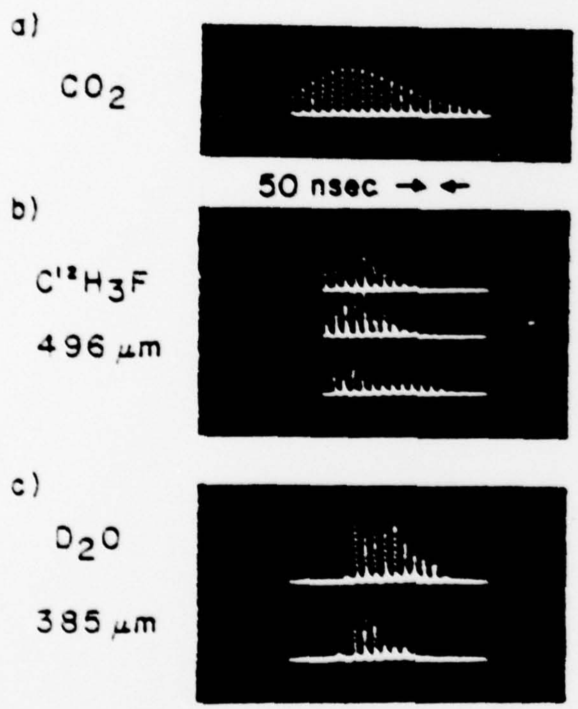


Fig. 2. Mode locked pulse trains from the system in Figure 1 with 1 torr in the FIR laser.

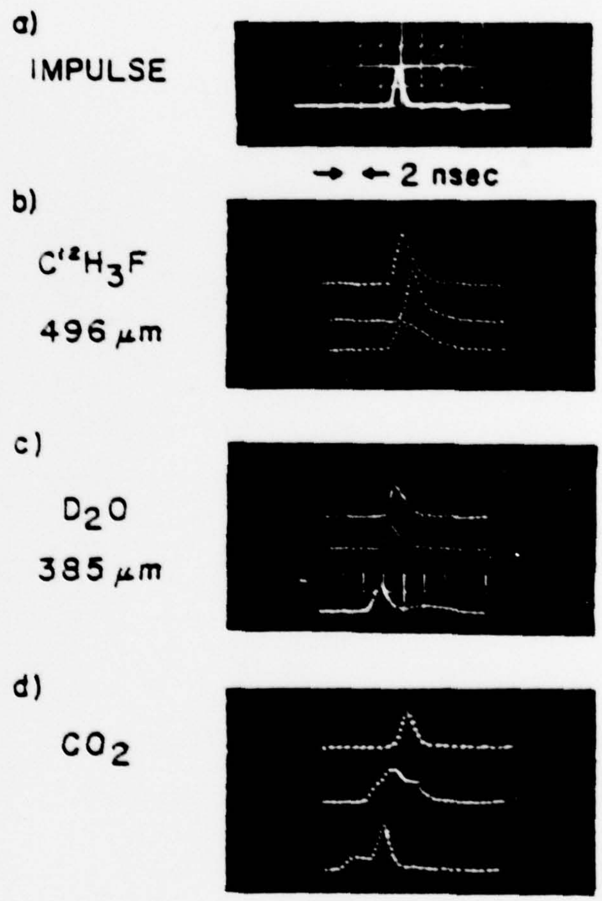


Fig. 3. Single pulses from the trains in Figure 2 showing full and partial mode locking. The impulse for a) was a 400 psec (FWHM) triangular pulse.

length of 3.6m. The entrance end of the 40 mm ID Pyrex waveguide was terminated with a Si-Brewster window whose interior surface was coated with a multi-layer IR dielectric mirror.[13] This mirror-window combination had an estimated FIR loss of $\lesssim$ 10% for wavelengths $\gtrsim$ 50 μm. In this configuration, the pump makes one round trip before being lost. A tungsten wire on nickel MOM diode, previously described,[14] was used to detect the FIR while a photon drag detector was used for IR detection.

An example of the resulting mode locked behavior is shown in Figure 2. Figure 2a) is the P9(20) CO_2 laser line with $\sim$ 5W RF drive to the modulator while Figure 2b) and Figure 2c) are the trains observed from $C^{12}H_3F$ at 496 μm and D2O at 385 μm, respectively, with $\sim$ 1 torr pressure in the FIR laser. (For reproduction purposes, the photographs were retouched with dots.) The peak FIR power was estimated to be between 1 and 10 kW for both lines.

On an expanded time scale, individual pulses can be resolved, examples of which are shown in Figure 3. The impulse response of the oscilloscope (Tektronix 485), Figure 3a), was 800 psec (FWHM) while the shortest FIR pulses ranged from 1.25-1.5 nsec which was the approximate range of the shortest pump pulses shown in Figure 3d). Incomplete mode-locking of the pump resulted in broadened pulses, examples of which are evident in this figure. We have also investigated the mode locked behavior in a simple single pass configuration and observed essentially the same behavior as with the cavity except that the FIR pulses were slightly shorter (1 nsec from a 7m cell).

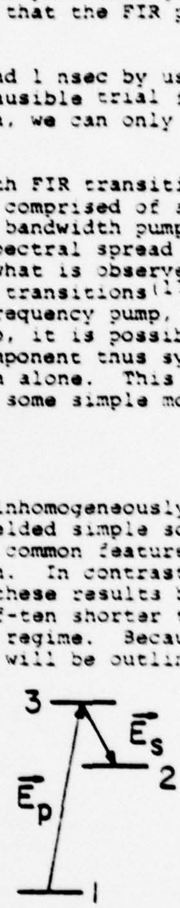
True FIR pulse widths were estimated to be between 500 psec and 1 nsec by using a simple convolution of the oscilloscope impulse response with various plausible trial functions. Thus since the actual pulse shape and detector speed are unknown, we can only claim that the true pulse widths are $<$ 1 nsec.

For the pressure conditions of Figure 3, the linewidth for both FIR transitions is 40 MHz. For the case of $C^{12}H_3F$, the IR transition, $\nu_3\ {}^QQ(12,K)$, is comprised of a K multiplet of which components K = 1-6 are thought excitable by the $\sim$ 1 GHz bandwidth pump.[15,16] The subsequent FIR emission from these multiplets would have a spectral spread of $\sim$ 500 MHz which could result in a mode-locked pulse of $\sim$ 2 nsec, close to what is observed. For D_2O, the absorption, $\nu_2\ 5_0 \rightarrow 4_0$, and emission, $4_0 \rightarrow 4_{-2}$, are isolated transitions[17,18] which would result in a mode locked pulse of $\sim$ 25 nsec with a single frequency pump, clearly much longer than what is observed. However with a multifrequency pump, it is possible to have a stimulated Stokes wave corresponding to each pump frequency component thus synthesizing a much greater bandwidth than is possible from a laser transition alone. This feature, thought present in both systems, can be further illustrated with some simple model treatments.

Theory

Previous treatments of mode locking in homogeneously[19] and inhomogeneously broadened[6] systems and of synchronously pumped systems [20] have yielded simple scaling relations which relate the pulse widths to the gain bandwidth, the common feature being that the pulse width is greater than or equal to the inverse bandwidth. In contrast, the experimental situations described here are not readily treated by these results because both the pumping pulse and emitted pulse are approximately a factor-of-ten shorter than the inverse linewidth placing the interaction in a transient or sub-T_2 regime. Because of this, alternate descriptions will be needed of which only the simplest will be outlined here.

First, in order to illustrate the stimulated Raman contribution to the overall gain bandwidth, we will consider a simple case of a CW mode locked pump interacting with a weak, tunable FIR probe wave. Since the interaction will be on a sub-T_2 time scale, a density matrix treatment as opposed to a rate equation treatment will be needed. Neglecting pump saturation and AC Stark shifts, the appropriate density matrix equations for the three-level system shown to the right are:



$$\partial\rho_{13}/\partial t = (i\Omega_{31} - 1/T_2)\rho_{13} + \vec{\mu}_{13}\cdot\vec{E}_p\rho_{11}/i\hbar \qquad (1)$$

$$\partial\rho_{12}/\partial t = (i\Omega_{21} - 1/T_2)\rho_{12} + \vec{\mu}_{23}\cdot\vec{E}_s\rho_{13}/i\hbar \qquad (2)$$

$$\partial\rho_{32}/\partial t = -(i\Omega_{32} + 1/T_2)\rho_{32} - \vec{\mu}_{31}\cdot\vec{E}_p\rho_{12}/i\hbar \qquad (3)$$

where the molecular transition frequency is defined in a positive sense as $\Omega_{ij} = (E_i - E_j)/\hbar$, $\vec{E}_p$ and $\vec{E}_s$ are the pump and Stokes fields and $\vec{\mu}_{ij}$ is the transition dipole moment. For simplicity, all T_2 are chosen equal to $1/\pi\Delta\nu_H$ where $\Delta\nu_H$ is the homogeneously broadened linewidth (FWHM) with a typical value of 40 MHz/torr. The Stokes gain is defined as

$$G_s = -\frac{4\hbar k_s}{\varepsilon_o |\vec{E}_s|^2} \, Im\left(\frac{\vec{\mu}_{23}\cdot\vec{E}_s}{\hbar}\,\rho_{32}\right) \tag{4}$$

With a mode locked pump of the form $\vec{E}_p = \sum_n \vec{E}_n \cos\omega_n t$ with n the pump logitudinal mode index, the Stokes gain at frequency ω_s and time t is found to be

$$G_s(\omega_s,t) = -\frac{\omega_s(\vec{\mu}_{23}\cdot\vec{\varepsilon}_s)^2 T_2\rho_{11}}{\hbar\varepsilon_o c} \, Im \sum_m \sum_n \frac{\Lambda_m\Lambda_n\, e^{i(x_m-x_n)t}}{L(x_n)L(x_n-y)L^*(y+x_m-x_n)} \tag{5}$$

where $\Lambda_n = \vec{\mu}_{13}\cdot\vec{E}_n/2\hbar$, $x_n = T_2(\Omega_{31}-\omega_n)$, $y = T_2(\Omega_{32}-\omega_s)$, $t = \tau/T_2$, $L(z) = z+i$ and $\vec{\varepsilon}_s$ is the polarization unit vector of the Stokes wave.

In Figure 4a) is shown a simple parabolic pump power spectra, approximating our situation, which would result in a mode locked pump of 1.2 nsec, close to what is observed. In Figures 4b) and c) are shown normalized Stokes gain profiles at t=0 for the pump in Figure 4a). The parameter D is defined as $D = (\Omega_{31}-\omega_o)T_2$, where ω_o is the pump center frequency, and is clearly the detuning from absorption line center. The two cases correspond to the $^QQ(12,2)$ absorption in $C^{12}H_3F$ (D=2) and the $5_0 \rightarrow 4_0$ absorption in D_2O (D=16) and illustrates the stimulated Raman emission contribution to the gain profile which can be further appreciated by comparison with a simple Lorentzian characteristic of a laser transition shown as the dots in Figure 4a).

Second, in order to estimate the Stokes pulse spectral width and spatial rate of growth, we employ a different set of approximations made primarily for analytical convenience. First we assume that the pumping pulses are tuned to line center (D=0) and are rectangular in time with a duration $\tau_p \ll T_2$ and peak amplitude Λ_p. Next, we neglect excitation to state $|3\rangle$ which means $\rho_{11} \sim$ constant and which also neglects any laser-like contribution to the Stokes gain. With these assumptions, the growth of the Stokes wave amplitude $\vec{E}_s$ in normalized units, $\Lambda_s = \vec{\mu}_{32}\cdot\vec{E}_s/2\hbar$, is governed by

$$\frac{\partial^3\Lambda_s}{\partial\tau^2\partial z} = G\tau\Lambda_s \qquad 0<\tau \le \tau_p \tag{6}$$

where retarded time τ is measured from the start of one of the pumping pulses and where $G = k_s(\vec{\mu}_{23}\cdot\vec{\varepsilon}_s)^2\rho_{11}\Lambda_p^2/4\varepsilon_o\hbar$ and is the equivalent steady state Stokes field gain divided by T_2 cubed. Taking the Laplace transform with respect to z, recasts Equation (6) into a standard form, the solutions of which are given by Airy functions or modified Bessel functions of fractional order.[21] The subsequent inversion of these functions results in a series which converges faster than the modified Bessel function series but can be approximated by the latter for not too large an argument. For large z, the approximate solution for $0 < \tau \le \tau_p$ is

$$\Lambda_s(x,\tau) \sim \frac{I_{-1/3}\left(\frac{2}{3}\sqrt{\alpha x}\right)}{x\left(\frac{1}{3}\sqrt{\alpha x}\right)^{1/3}} \sim \frac{e^{\frac{2}{3}\sqrt{\alpha x}}}{\alpha^{\frac{5}{12}}\,x^{\frac{17}{12}}} \tag{7}$$

where $\alpha = LG\tau^3$ and $x = z/L$, with a maximum interaction length L. The parameter α is the normal Stokes field gain coefficient with T_2 replaced by τ, the latter controlling the linewidths in this case. The basic result, Equation (7), can be compared with the equivalent steady state field growth given by

a) Synthetic power spectra approximating the pump, with X a normalized detuning from linecenter.

b) Stokes gain profile for the pump in a) with Y a normalized detuning. D is the normalized mismatch between the pump center frequency and the absorption. G_s is calculated from Equation 5. Y = 0 is centered over X = D.

c) Same as b) except with a bigger mismatch. For the conditions of Figure 1, 1 unit = 20 MHz.

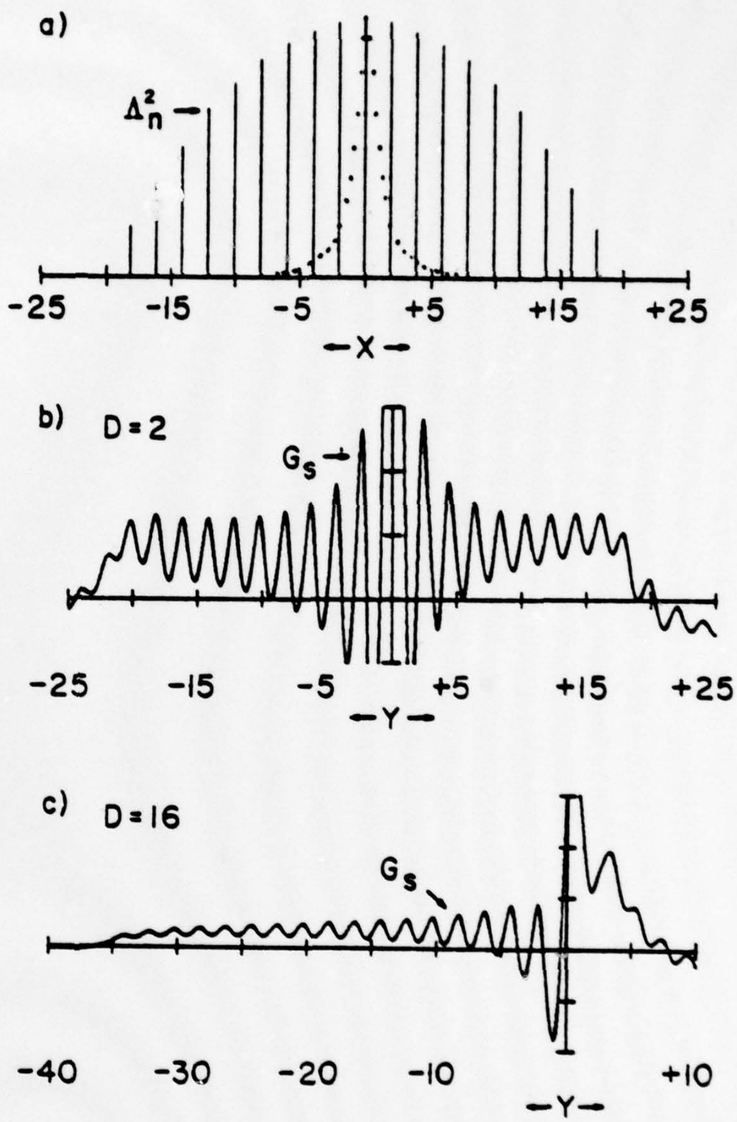


Fig. 3.

$$A_s(x) \sim e^{GT_2^3 Lx} \tag{8}$$

and growth in a transient regime for D >> 1 given by

$$A_s(x,\tau) \sim \frac{e^{\sqrt{g\tau xL}}}{(g\tau xL)^{1/4}} \qquad 0<\tau \le \tau_p \tag{9}$$

with gT_2, the equivalent steady-state gain.(5,22,23) The key features which thus emerge for the transient case are that the Stokes fields grows spatially as $e^{\sqrt{x}}$, which is considerably slower than the normal form of Equation (8), and that because of the exponential dominance

of Equation (7), with time dependence $e^{\tau^{3/2}}$, the Stokes pulse may be temporally narrower (spectrally broader) than the pump, contracting (broadening) further as it propagates. Similar features have been discussed before in conjunction with the normal Raman situation (D>>1) under transient pumping[22,23] and have been qualitatively observed in these experiments in a long single pass amplifier.

The major conclusions we draw from these analyses are that Stokes gain corresponding to each frequency component of the pump is significant, that the growth of the Stokes wave is 'lethargic' in the sense that the material is attempting to respond on a sub-T_2 time scale and that the temporal duration of the Stokes wave may be significantly narrower than that of the pumping pulse. The presence of pump saturation with a resulting FIR laser-like gain, and pump depletion are not expected to modify these conclusions significantly but may set explicit limits on them. These and other features will be explored in the future.

Other Approaches

There are two other approaches worthy of mention at this point, a self compression in high gain amplifier systems and an external shuttering caused by free carrier absorption. The latter is illustrated conceptually by considering a high purity semiconductor oriented at Brewster's angle with respect to a FIR source. Under these conditions, the FIR wave suffers no reflection or attenuation in transmission. If the surface of the semiconductor is subsequently illuminated with light with a photon energy greater than the bandgap energy, electron-hole pairs will be created near the surface, causing absorption in the FIR. For a sufficiently high carrier density, a metallic-like behavior is created resulting in reflection in the FIR. A system of two semiconductors, one used in reflection modulation and the other in transmission modulation, can act as a shutter producing single pulses. Experiments illustrating these features have been reported in the infrared[2,3] and FIR.[24]

The main virtue of the approach is speed, with shuttering rise times determined by the 'writing' pulse duration which can be in the psec range. However, calculations for Si with FIR wavelengths $\sim$ 500 μm indicate that 'writing' energy densities of $\sim 10^{-5}$ (10^{-4}) j/cm^2 are needed to create significant absorption (reflection) which corresponds to psec pulse powers of 10(100) MW/cm^2.[24] This large required writing energy or power density is the main disadvantage of the method.

The two self compression schemes, swept-gain superradiance[5,25] and backward wave stimulated Raman emission,[4] are similar in that they may occur in optically pumped systems and have 'steady-state' or stable output pulses described by

$$I(\tau) = \frac{B}{\tau_s^2} \mathrm{Sech}^2\left(\frac{\tau}{\tau_s} - \eta\right) \tag{10}$$

where B and η are some constants and $\tau_s = T_2\kappa/G_F$ where κ is a field loss coefficient and G_F is a gain coefficient. Since the pulse width from Equation (10) is $\Delta\tau = 1.76\ \tau_s$, then for high gain to loss ratios (G_F/κ), $\Delta\tau << T_2$. The major difficulty with these approaches is that a fully contracted (temporally narrow) pulse occurs in an interaction distance $z \sim 10\kappa^{-1}$, which, since G_F is bounded in most systems, may be very long, $\sim$ 10's of meters.

Conclusions

We have demonstrated that it is possible to generate FIR pulses that are at least 20 times shorter than the inverse linewidth, a fact attributed to an effective inhomogeneous broadening in molecular systems with a K multiplet structure or to stimulated Raman emission from each frequency component present in the pump; a combination of both may also exist. Similar results were achieved in a synchronously pumped cavity and in a single pass cell; the similarity is probably due to our inability to fully resolve the pulses because of the speed limitations of the detection system. In fact new correlation techniques will have to be developed before better estimates can be made since the present speeds, which are thought to be $\sim$ 500 psec, are at the current technologic limit of single pulse detection. Of the four approaches discussed, free-carrier switching will probably result in the shortest pulses, but with an intensity that may be lower than the other three. In particular, the mode-locked pumped system, which already produces pulses with powers which are about the Manley-Rowe limit ($\sim$2%) of the average pumping power (200 kW) but $\sim$ 20 times lower than the limit for the peak pump power (5MW) used, has yet to be fully exploited in terms of the potentials of the approach. For example, recent experiments have demonstrated tunable FIR Raman emission over $\sim$ 5 cm^{-1} range in optically pumped HCl and HF which could result in $\sim$ 10 psec FIR pulses with the use of a mode locked pump.[26,27] Thus further improvements in pulse duration and conversion efficiency may be possible.

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